

# Scaling Models for Thermodynamic Properties of HFC 134a and HFC 143a on the Coexistence Curve

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## Abstract

The chief aim of the work was to obtain scaling forms for the properties  $F$  (the densities  $\rho_l$  and  $\rho_g$ , the saturation pressure  $P_s, dP_s/dT$  and the specific enthalpy of vaporization  $h$ ) of HFC 134a and HFC 143a along the coexistence curve (CC) including a wide region around the critical point. A methodical part dealt with a general equation of a property  $F(\rho_l, \rho_g, P_s)$  that had a scaling part  $F_{scal}$  and a regular one  $F_r$ . The form of  $F_{scal}$  followed to the scaling theory (ST) and depended on the relative distance  $\tau$  from  $T_c$ , critical exponents  $(\alpha, \beta, (\Delta_i))$  and amplitudes,  $(B_{si}), (B_{di})$ .

Experimental data on  $P_s, \rho_l, \rho_g$  and  $h$  were analyzed. Reliable results were involved in the input data sets those covered the region from the triple point temperature  $T_{tr}$  to  $T_c$ . Among them there were new results of Magee [1,2] and Yata [3] for HFC134a. Analogous new data [4-5] were chosen for the input data sets prepared for HFC 143a. New data are placed in the interval  $0.005 < \tau < 0.25$ . A statistical routine and some criterions were elaborated for the task. The form of scaling function  $\psi_{l,g}$  was used to analyze  $\rho_l(T)$  and  $\rho_g(T)$ . A system of relations  $P_s(T), \rho_l(T), \rho_g(T)$  and  $h(T)$  was produced and proposed for joint calculation of  $P, \rho, h, T$  - data in a wide temperature region up to  $T_c$ . The equations,  $\rho_l(T), \rho_g(T)$  a.o., used effective values of parameters,  $T_c, \rho_c, \alpha, \beta, B_{s0}, B_{d0}$ , determined with the help of the routine.

The relations represented reliable measured points including new data in limits of experimental errors. They can be helpful to calculate properties in a wide region of the critical point,  $0.005 < \tau < 0.1$  were known tables [7 - 10 a. o.] report only a few data.

**Keywords:** HFC 134a; HFC 143a; Thermodynamic properties; Scaling equation; Saturation pressure; Density of liquid, Density of vapor; Coexisting curve.

## 1. Introduction

A methodical part of the investigation deals with a general equation of a property  $F$  that has the structure

$$F(T) = F_{scal}(\tau, \beta, \alpha, (\Delta_i), (B_i)) + F_r(\tau), \quad (1.1)$$

where  $F_{scal}$  – a scaling part,  $F_r$  – a regular part.

$F_{scal}$  has the form that follows to ST, is discussed in [11,12] and includes three addends in a general case.  $F_{scal}$  of the order parameter  $f_s$  and the diameter of CC  $f_d$  looks like

$$f_s = (\rho_l - \rho_g)(2\rho_c)^{-1} = B_{s0}\tau^\beta + B_{s1}\tau^{\beta+\Delta_1} + B_{s2}\tau^{\beta+\Delta_2}, \quad (1.2)$$

$$f_d = (\rho_l + \rho_g)(2\rho_c)^{-1} - 1 = B_{d0}\tau^{1-\alpha} + B_{d1}\tau^{1-\alpha+\Delta_1} + B_{d2}\tau^{1-\alpha+\Delta_2}. \quad (1.3)$$

The first addend in (1.2) (1.3) represents the asymptotic component, the second and the third terms give non asymptotic components (the first and the second correction terms with correction exponents  $\Delta_1$  and  $\Delta_2$ ).

Analytic  $\rho(T)$  and  $\rho_g(T)$  equations can be determined with the help of known (1.2,1.3) as

$$\rho_l = (f_d + f_s + 1) \rho_c, \quad \rho_g = (f_d - f_s + 1) \rho_c. \quad (1.4)$$

Equations (1.2,1.3) are named as Model 1 that consists of  $F_{scal}$  entirely. Its approximation quality is characterized by individual and RMS deviations of experimental ( $\rho_{g\ exp\ k}, \rho_{l\ exp\ k}$ ) values from those calculated with equations (1.4)

$$\begin{aligned} \delta\rho_{gk} &= 100 (\rho_{g\ exp\ k} - \rho_{gk}) / \rho_{gk}, & S_g &= (\sum \delta\rho_{gk}^2 / N)^{0.5}, \\ \delta\rho_{lk} &= 100 (\rho_{l\ exp\ k} - \rho_{lk}) / \rho_{lk}, & S_l &= (\sum \delta\rho_{lk}^2 / N)^{0.5}, \end{aligned} \quad (1.5)$$

Model 1 can be considered as  $Y = (f_s, f_d)$  that includes critical characteristics  $D = (\rho_c, T_c, \alpha, \beta, (\Delta_i))$  and coefficients  $C = ((B_{si}), (B_{di}))$ . Values of  $C$  are to be determined by an approximation of the input data set ( $Y_{exp\ k}, \tau_k$ ). If  $D$  are considered as the parameters fixed and known (taken from literature sources) then  $Y = f(D, C, \tau)$  is a linear function of  $C$ . In the case it is possible to calculate  $C$  using a weighted least-squares analysis (LSQA) and a single criterion – a minim of the functional  $\Phi(C, D)$

$$\Phi(C,D) = \sum_{k=1}^N w_k (Y_{exp\ k} - f(D,C, \tau_k))^2 / N = \min. \quad (1.6)$$

Routine 1 is elaborated for  $C$  calculation and has the following steps: i) to consider  $D$  as the parameters fixed and to take from literature sources; ii) to form a sum of squares,  $\Phi(C,D)$ , for the input data set; iii) to calculate a realization  $C$ , that is numerical values of  $C$ , using a weighted least-squares analysis (LSQA) and a single criterion – a minim of  $\Phi(C,D)$ ; iii) to estimate a quality of the approximation that is to calculate individual and RMS deviations .

Model 1 let us represent experimental data in a working interval  $\Delta \tau_w = 0.15$  of  $T_c$  ( $T_c > T > T_w$ ,  $\Delta \tau_w = 1 - T_w/T_c$ ) for different substances [11,12,13]. Deviations,  $S_g, S_l$ , were small and no systematic deviations were realized. Our practice showed if  $T_w$  decreases and  $\Delta \tau_w > 0.15$  then RMS deviations,  $S_g, S_l$ , increase and remarkable systematic errors appear.

To represent  $\rho_b, \rho_g, T$  – data in  $\Delta \tau_w > 0.15$  we have elaborated a combined model – Model 2 [12,13] that includes  $F_{scal}$  and additional  $F_r(\tau)$  in comparison with Model 1

$$f_s = B_{s0} \tau^\beta + B_{s1} \tau^{\beta+\Delta 1} + B_{s2} \tau^{\beta+\Delta 2} + B_{s3} \tau^2 + B_{s4} \tau^3, \quad (1.7)$$

$$f_d = B_{d0} \tau^{1-\alpha} + B_{d1} \tau^{1-\alpha+\Delta 1} + B_{d2} \tau^{1-\alpha+\Delta 2} + B_{d3} \tau^2 + B_{d4} \tau^3. \quad (1.8)$$

$F_r(\tau)$  is involved with the aim to reduce systematic deviations mentioned for Model 1 under  $\tau_w > 0.15$ . Model 2 is to have the following characteristics: 1) it approximates  $\rho_b, \rho_g, T$  – data in  $\Delta \tau_w > 0.15$  with acceptable deviations,  $S_g, S_l$ , 2) at the same time its scaling part delivers acceptable deviations,  $S_g, S_l$ , for points in  $\Delta \tau_w = 0.15$  when  $F_{scal}$  is considered as Model 1 and used to calculate deviations of  $\rho_b, \rho_g, T$  – data from  $F_{scal}$  in  $\Delta \tau_w = 0.15$ .

One more property is considered in ST and involved in the work: the scaling function  $\psi_{l,g}$  that can be expressed in the form

$$\psi_{l,g} = |(\rho_{l,g} - \rho_c)(2\rho_c \tau^\beta)^{-1}| = |\pm B_{s0} + B_{d0} \tau^{1-\alpha-\beta} \pm B_{s1} \tau^{\Delta 1-\beta} + B_{d1} \tau^{1-\alpha+\Delta 1-\beta} + \dots|, \quad (1.9)$$

where the upper (plus) sign of terms corresponds to the liquid branch ( $l$ ), the lower (minus) corresponds to the vapor branch ( $g$ ) of  $\psi_{l,g}$ .

The scaling part of the equation  $P_s(T)$  is written as

$$\ln(P_s/P_c)_{scal} = B_{pi} \tau^{2-\alpha} + B_{pi+1} \tau^{2-\alpha+\Delta l}. \quad (1.10)$$

The form (1.8) follows to the next degree law in the asymptotic region  $\Delta\tau_w$

$$dP_s^2/dT^2 \sim \tau^{-\alpha}. \quad (1.11)$$

Several data sources and criterions are used in the work to establish the models and to determine critical exponents and parameters and amplitudes of the models.

## 2. Density: models, criterions and routines

A preliminary treaty of experimental and tabulated  $\rho_l(T)$  and  $\rho_g(T)$  –data for HFC 134a ( $\approx 400$  points) let us select an input  $\rho_b \rho_g, T$  – data set that consists of  $\approx 200$  points including data from [2,3,14-17] those were not used in the known correlations. They cover a region from  $T_{lr} = 169.85$  to  $T_c$  ( $\Delta\tau_w \approx 0.5$ ). It is important to remark that the investigation [3] has given  $\rho_b \rho_g, T$  – values those are got with a refractive index technique in the interval  $\Delta T_w \approx 5$  K where a big scattering ( $\pm 4\%$ ) of points existed. The results [2] gave an accurate information in  $\Delta\tau_w \approx 0.5$  including data near  $T_{lr}$ . An analog input  $\rho_b \rho_g, T$  – data set is formed for HFC 143a including data [4,5] those are placed in  $\Delta\tau_w = 0.25$ .

Routine 1 gives an opportunity to examine a group of realizations ( $Y = f(D_j, C_j, \tau)$ ,  $j = 1 \dots K$ ). Criterion (1.6) shows that  $\Phi(C, D)$   $S_{l,g}(D)$  are functions of parameters  $D$ . If Routine 1 is used at the first time then the following numerical values appear parameters,  $D = D_1$ , a realization,  $C = C_1$ , RMS deviations,  $S_{l,g}(D_1)$ . If one of the parameters  $D_1$  is shifted (for example  $\rho_c$  can be shifted on  $\Delta\rho_c$  in limits of an experimental estimation  $\pm \Delta\rho_c$ ) then Routine 1 can be used at the second time and the second set,  $D_2, C_2, S_{l,g}(D_2)$ , can be got. Many numerical variants (realizations ( $C_j$ )) of Models 1,2 and according deviation sets ( $S_{l,g}(D_j)$ ) were got, among them there were such numerical dependences as  $S_{l,g}(\rho_c)$ ,  $S_{l,g}(T_c)$ ,  $S_{l,g}(\beta)$  in a wide region of  $S_l^{min}$  and

$S_g^{min}$ . Our analyses estimated some general features of the realisations: i) values of  $S_g$  and  $S_l$  did not coincide ( $S_l < S_g$ ), the minimal values were found between them ( $S_l^{min} = 0.22\%$  and  $S_g^{min} = 0.65\%$  for HFC 134a points related to  $\Delta\tau_w = 0.25$ , ii) the leading amplitudes ( $B_{s0}, B_{d0}$ ) deviated greatly from theoretical values ( $B_{s0} \approx 2$  and  $0 < B_{d0} < 1$ ) and from middle calculated values ( $B_{s0}, B_{d0}$ ), iii) it was impossible to find a variant  $C_j$  that delivered  $S_l = S_l^{min}$  and  $S_g = S_g^{min}$  at once. The last circumstance was one of the reasons to look for an optimal Models 1,2 ( $Y = f(D_{opt}, C_{opt}, \tau)$ ) with an optimal realization  $C_{opt}$  that deliver compromise values of  $S_{l,g}(D_{opt})$ . It was admitted that realization  $Y = f(D_{opt}, C_{opt}, \tau)$  is optimal dew to the approximation quality when  $Y$  delivers RMS deviations those follow to the compromise criterions

$$S_l(D_j) - S_l^{min} < \varepsilon_1, \quad S_g(D_j) - S_g^{min} < \varepsilon_2, \quad (2.1)$$

where  $\varepsilon_1$  and  $\varepsilon_2$  – some limits.

Our practice and analysis of a realizations, ( $C_j$ ), have shown that more criterions have to be involved for the purpose to choose an optimal variant from realizations ( $C_j$ ) those satisfied criterions (2.1).

Investigations [1,12,13] estimated that the scaling function  $\psi_{l,g}$  can be used as an additional criterion.  $\psi_{l,g}$  is written in a narrow interval  $\tau_w \approx 0.01$  as two terms expression that includes only leading addends with the asymptotic exponents

$$\psi_{l,g} \approx |\pm B_{s0} + B_{d0} \tau^{1-\alpha-\beta}|. \quad (2.4)$$

A numerical form (2.4) of  $\psi_{l,g}$  was found and analysed for several liquids ( $H_2O, Ne, N_2, HFC$  134a, HFC 143a a. o.). Due to (2.4) the next features of variants ( $Y = f(D_j, C_j, \tau)$ ): i)  $\psi_{l,g}$  is symmetric to the amplitude  $B_{s0}$  for liquid and vapour branches, ii) the plot of  $\psi_{l,g}$  versus  $\tau^{1-\alpha-\beta}$  is liner and converge symmetrically to  $B_{s0}$ , iii) corresponding Model 0 as  $Y = f(D, C = B_{s0}, B_{d0}, \tau)$  can be built, the asymptotic Model 0 are in a satisfied agreement with experimental points related to

$\tau_w \approx 0.01$  and deviations,  $S_b, S_g$ , from Model 0 are close to criterions (2.3). An illustration of  $\psi_{l,g}$  for HFC 134a is given in Fig. 1.

Two methods of  $D$  chose were examined for the task. Due to the first one  $D$  components are taken as theoretical or experimental values and not varied during a treaty of the input data set. For example the exponents are taken in [6,16,17] as theoretical data  $\beta=0.325$ ,  $\alpha=0.1085$  and  $\Delta = 0.5$ . We have used the way for  $(\Delta_i)$  determination and chosen  $\Delta_1 = \Delta = 0.5$  and  $\Delta_2 = 2\Delta$  as theoretical values. The second method [11,12,13] considers  $D = (\rho_c, T_c, \alpha, \beta, B_{s0}, B_{d0})$  as fixed parameters but  $D$  is known in a region of possible deviations  $(\pm \Delta D)$ . The method let us shift  $D$  in the limits  $(\pm \Delta D)$  corresponding to some criterions with the aim to reach a satisfy correlation of Models 1,2 with the input data set. We underline that leading amplitudes,  $B_{s0}, B_{d0}$ , are also included in  $D$  and they do not take part in LSQA when amplitudes  $C$  are determined. This schema has been accepted in the work.

The middle value of  $D$  components and a region of possible deviation  $(\pm \Delta_{\rho_c}, \pm \Delta_{T_c}, \pm \Delta_{\beta}$  a.o.) were found previously. The start values of the exponents  $(\alpha, \beta)$  were chosen as  $\alpha=0.1085$  and  $\beta=0.325$ . The characteristics were combined in a group  $D_1 = (\rho_c, T_c, \alpha, \beta, \Delta, B_{s0}, B_{d0})$  as a start data.

Routine 2 was elaborated using criterions (2.3,2.4). It consisted of several steps: i) to take  $D_1$  as start fixed parameters, ii) to calculate the realization  $C_1$  employing weighted LSQA, iii) to determine the criterions  $(S_l, S_g, \psi_{l,g})$  and to analyse them; if the analysis showed that criterions  $(S_l, S_g, \psi_{l,g})$  satisfied conditions (2.3,2.4) then the routine was to finish. On opposite site, a new fixed parameter (for example, it was  $\rho_c$ ) was to be chosen ( $\rho_c$  was shifted on  $0.1 \Delta_{\rho_c}$ ),  $D_2$  was formed and treaty was continued (steps ii,iii) to get next realisation  $C_2$ .

Routine 2 let us get Models 1,2 those had effective parameters,  $D_{opt}, C_{opt}$ , and were agree with conditions (2.3,2.4). The routine was used to build Model 2 approximated  $\rho_l, \rho_g, T$  –points of HFC 134a in  $\tau_w \approx 0.5$ . The limits,  $\varepsilon_1, \varepsilon_2$ , were determined as  $\varepsilon_1 = 0.1\%$  and  $\varepsilon_2 = 0.2\%$ . Numerical

parameters of Model 2 are placed in table 1. Model 2 was fitted to the input data set with acceptable accuracy in  $\tau_w \approx 0.2$  (gas phase) and  $\tau_w \approx 0.5$  (liquid phase).

**Table 1.** Parameters of Model 2 for HFC 134a.

$\rho_c / \text{kg/m}^3$	$T_c / \text{K}$	$\alpha$	$\beta$	$\Delta$
510.5	374.105	0.1509	0.34942	0.5
$B_{s0}$	$B_{s1}$	$B_{s2}$	$B_{s3}$	$B_{s4}$
2.11	-0.044561	-0.426163	-0.039283	0.474428
$B_{d0}$	$B_{d1}$	$B_{d2}$	$B_{d3}$	$B_{d4}$
0.525	0.853378	-0.42822	0.029534	-0.008737

**Table 2.** Parameters of Model 2 for HFC 143a.

$\rho_c / \text{kg/m}^3$	$T_c / \text{K}$	$\alpha$	$\beta$	$\Delta$
430.66	345.815	0.22	0.3515	0.5
$B_{s0}$	$B_{s1}$	$B_{s2}$	$B_{s3}$	$B_{s4}$
2.077	1.793437	-1.364958	+0.199057	0.730117
$B_{d0}$	$B_{d1}$	$B_{d2}$	$B_{d3}$	$B_{d4}$
0.223	0.084649	-0.5101183	+0.210395	0.717498

### 3. Saturation pressure and specific heat of vaporization

An expression of  $P_s(T)$  was chosen in the form:

$$\ln(P_s/P_c) = B_{p0} (1 - 1/t) + B_{p1}\tau + B_{p2}\tau^{2-\alpha} + B_{p3}\tau^{2-\alpha+\Delta} + B_{p4}\tau^3 + B_{p5}\tau^5, \quad (3.1)$$

where,  $(B_{pi})$  - adjustable coefficients.

Values of  $B_p(B_{pi})$  were determined under a statistical fitting of  $P_s, T$ –data. Approximation criterions were connected with deviations of experimental data from equation (3.1) and expressed as

$$\delta P_i = 100 (P_{exp\ i} - P_i) / P_i, \quad S_p = (\sum \delta P_i^2 / N)^{0.5}, \quad (3.2)$$

where  $P_{exp\ i}, P_i$  – experimental and calculated values of  $P$  in  $i$  – point,  $S_p$  – a relative RMS deviation,  $N$  number of points considered.

One more criterion was taken into consideration – a relative RMS deviation ( $S_h$ ) of experimental  $h, T$  – data. An analysis of experimental and tabulated  $P, T$  – data let us select an input data set ( $\approx 100$  points including new data [2,14,15,18]). The input data set covered a region  $\Delta T_w = 205$  K from  $T_{tr} = 169.85$  to  $T_c$ .

A routine of  $B_p$  determination was elaborated. As in the case of the density treaty, it consisted of analogous steps: i) to take  $D_1 (P_c, T_c, \alpha, \Delta, B_{p0})$  as start fixed parameters, ii) to calculate the realization  $(B_{pi})_1$  employing a minimization of  $\Phi(C, Y_{exp\ i}, Y_i, \tau_i, w_i)$ , where  $Y = \ln(P/P_c)$ , and iii) to determine criterions ( $S_p, S_h$ ) and to analyse them. The next step is analogous to the step that takes place in Routine 2. Values of  $(B_{gi})$  were calculated and shown in table 3.

**Table 3.** Parameters of saturation pressure equation for HFC 134a

$P_c/\text{MPa}$	$T_c/\text{K}$	$\alpha$	$\Delta$	$B_{p0}$	$B_{p1}$	$B_{p2}$	$B_{p3}$	$B_{p4}$	$B_{p5}$
4.0502	374.105	0.1509	0.5	9.5	1.90843	7.63554	-11.35781	11.72233	-10.83595

**Table 4.** Parameters of saturation pressure equation for HFC 143a

$P_c/\text{MPa}$	$T_c/\text{K}$	$\alpha$	$\Delta$	$B_{p0}$	$B_{p1}$	$B_{p2}$	$B_{p3}$	$B_{p4}$	$B_{p5}$
3.761	345.815	0.22	0.5	8.50	1.17781	5.67519	-7.64488	8.55837	-9.18576

The specific enthalpy of vaporization was represented with a known formula

$$h = \frac{dP_s}{dT} T \left( \frac{1}{\rho_s} - \frac{1}{\rho_l} \right), \quad (3.3)$$

where  $dP_s/dT$  – the derivative determined with the help of (3.1),  $\rho_g, \rho_l$  – determined with the help of (1.7,1.8).

#### 4. Conclusion

The analysis of realizations, an optimal variant of Model 2 and results of a comparison with literature data allows us to make an output that the scaling model (1.7,1.8) can be useful to approximate experimental  $\rho_l, \rho_g$  – data on a line of phase equilibrium including a broad



neighborhood of the critical point. Numerical variants of Model 2 for HFC134a and HFC143a can improve the known reference data [7 - 10] in the region  $0.005 < \tau < 0.05$  were known tables [7 - 10 a. o.] report only a few data. The equations,  $\rho_l(T), \rho_g(T)$  a. o., used effective values of parameters,  $T_c, \rho_c, \alpha, \beta, B_{s0}, B_{d0}$ , determined with the help of the routines elaborated.

$P_s(T)$  equation (3.1) represents experimental data with low errors. RMS deviation of the input data set  $S_p$  for HFC 134a is determine as  $S_p = 0.04\%$ . Deviations of  $P_s, T$  – data [2] for HFC 134a are placed in the limits  $\delta P = \pm 0.08\%$  in the interval 169.85...250 K.  $S_p$  of data [14,15] is determined as  $S_p = 0.72\%$ . Remarkable deviations,  $\delta P = -4.2 - 7.4\%$ , is related to the interval 169,85...190 K, at higher  $T$  the points [14,15] have deviations  $\delta P = -0.23...0.32\%$ . A satisfied agreement is got for data (Goodwin at al, 1993,  $\delta P = -0.02 - 0.06\%$ , Baehr at al, 1991,  $\delta P = -0.02 - 0.03\%$ ).

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#### *List of symbols*

$T$  = temperature

$P$  = pressure

$\rho$  = density

$P_s$  = saturated pressure

$g, l, c$  = indexes to mark the vapor and liquid phases on CC and a value in the critical point

$\alpha, \beta, (\Delta_i)$  = critical exponents

$\tau = 1 - T/T_c$  = relative distance of temperature from  $T_c$ ,

$(B_{si}), (B_{di}), (B_{pi})$  = amplitudes

$\delta\rho_{gk}, \delta\rho_{lk}$  = relative deviations of the vapour and liquid densities in  $k$  – point

$\rho_{g\exp k}, \rho_{l\exp k}$  = experimental values of the the densities

$\rho_{gk}, \rho_{lk}$  = values of the vapour and liquid densities calculated with (1.4)

$N$  = the number of points in the input data set

$S_g, S_l$  = relative RMS deviations of vapour and liquid densities.

$w_k$  = weight coefficient for  $k$  – point

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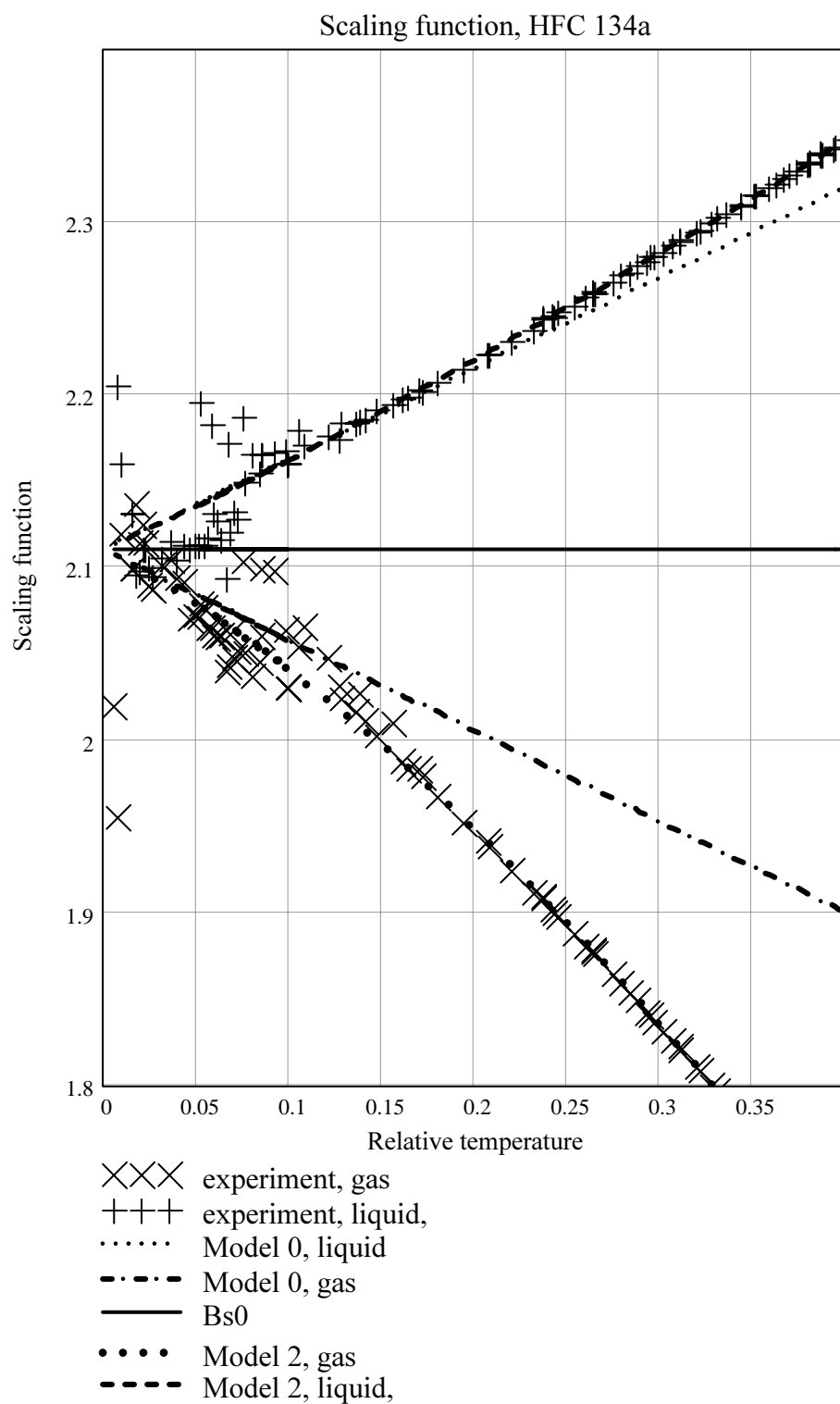


Fig. 1. The scaling functions  $\psi_{l,g}$  of HFC 134a.  
 Values are determined from 1) the experimental data, 2) Models 0,2;  
 Bs0 – leading amplitude  $B_{s0}$ ; relative temperature –  $\tau^{1-\alpha-\beta} = (1 - T/T_c)^{1-\alpha-\beta}$